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## Request for grant of a patent

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| | | |
|---|---|---|
| 1. Your reference | 11050P4 GB/EC | |
| 2. Patent application number *(The Patent Office will fill in this part)* | 0214999.5 | 28JUN02 E729543-1 D02903 P01/7700 0.00-0214999.5 |
| 3. Full name, address and postcode of the or of each applicant *(underline all surnames)* | Reckitt Benckiser N.V.<br>Kantoorgebouw De Appelaer<br>Fruittuinen 2-12<br>2132 NZ Hoofddorp<br>NETHERLANDS | 28 JUN 2002 |
| Patents ADP number *(if you know it)* | 07921075005 | |
| If the applicant is a corporate body, give the country/state of its incorporation | Netherlands | |
| 4 Title of the invention | Detergent Composition | |
| 5. Name of your agent *(if you have one)* | Craig M Bowers | |
| "Address for service" in the United Kingdom to which all correspondence should be sent *(including the postcode)* | Reckitt Benckiser plc<br>Group Patents Department<br>Dansom Lane<br>HULL<br>HU8 7DS<br>UNITED KINGDOM | |
| Patents ADP number *(if you know it)* | 07799521001 | |

| | | | |
|---|---|---|---|
| 6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and *(if you know it)* the or each application number | Country | Priority application number *(if you know it)* | Date of filing |
| 7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application | Number of earlier application | | Date of filing *(day / month / year)* |

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? *(Answer 'Yes' if:*
*a) any applicant named in part 3 is not an inventor, or*
*b) there is an inventor who is not named as an applicant, or*
*c) any named applicant is a corporate body.*
*See note (d))*

Yes

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| Continuation sheets of this form | |
| Description | 12 |
| Claim(s) | 4 |
| Abstract | 1 |
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10. If you are also filing any of the following, state how many against each item.

| | |
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| Priority documents | |
| Translations of priority documents | |
| Statement of inventorship and right to grant of a patent *(Patents Form 7/77)* | |
| Request for preliminary examination and search *(Patents Form 9/77)* | One |
| Request for substantive examination *(Patents Form 10/77)* | One |
| Any other documents *(please specify)* | FS2 |

11. I/We request the grant of a patent on the basis of this application.

Signature

Craig M.Bowers

Date

28 June 2002

12. Name and daytime telephone number of Person to contact in the United Kingdom

Craig Bowers (01482) 583730

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DUPLICATE



# DETERGENT COMPOSITION

The present invention relates to a detergent composition, more particularly to a detergent composition comprising an enzyme partially disposed within particles in a gel, wherein the particles have a migration speed of less than 1 centimetre / month.

Enzymes find increasing use in detergents as a result of their ability to aid the removal of organic soils and stains from domestic articles. Enzymes are especially useful in the dispersion of food stains on clothing and cooking/eating utensils. Typical enzymes employed in this fashion include proteases to aid the removal of proteins and amylases, which act upon starch.

Unfortunately enzymes in detergent formulations, especially water-based formulations normally exhibit very poor stability. This problem is especially true at elevated temperatures and under the presence of UV light. Attempts to address this disadvantage in water based gel detergent formulations have included the application of known technologies such as increasing the ionic strength of a water based gel containing enzymes or by adding stabilising agents to the gel. However, a substantial deterioration of enzymes is still observed.

We have now found that the enzyme stability in such systems can be increased to a surprisingly high level when the enzymes are partially encapsulated and the so formed particles are then added to a gel in which the particles have limited mobility.

According to the present invention there is provided a detergent composition comprising an enzyme which is at least partially disposed within water-soluble particles in a gel, the particles comprising a water-soluble encapsulating agent, wherein the particles have a migration speed in the gel of less than one centimetre per month.

It has been found that as a result of the low motility of the particles, the particles once dispersed in the gel, remain dispersed therein, even after long periods of storage. Thus the problems of particle interaction and damage, as a result of particle congregation at or near an upper or lower portion of the gel are overcome.

Additionally as the particles remain evenly dispersed, even over prolonged periods of storage, the user can be sure when measuring / dispensing an amount of the detergent gel, that it contains the correct (rather than an excessive or insufficient) amount of particles (and associated enzyme). Furthermore the correct level of dispense may be achieved without the need to shake or otherwise agitate the gel which could otherwise cause detrimental particle deterioration.

Also, even though the enzyme is protected in storage in the particles, the particles are quickly disintegrated in use in a wash liquor (by virtue of the water-soluble encapsulating agent), thus allowing the enzyme to perform its function without delay.

Thus the current invention has been found to provide an enzyme containing detergent gel composition which displays surprisingly good enzyme stability during storage, whilst also ensuring a quick and efficient release of enzyme in use.

Preferably the migration speed of the particles is less than 0.7 cm per month and most preferably less than 0.4 cm per month.

Without wishing to be bound by theory the migration speed of the particles may be measured by the following preferred, yet non-limiting method.

The particles are dispersed in the gel and the gel is placed in a closed glass bottle (capacity 50ml, width 3.5cm). A picture is taken (Canon Powershot 30S camera, with the distance lens-bottle being 50cm). The bottle is stored for 30 days at 25°C. A second picture is taken from the glass bottle and the locations of the particles are compared. Changes of location (Migration distance of particle on picture = $D_p$) are recorded in cm. The migration distance $D_r$ of an individual particle is determined

according to the following formula, which overcomes any parallax error introduced by the picture taking process.

$D_r = H_r \times D_p / H_p$

$H_p$ = Bottle height on picture
$H_r$ = Real bottle height
$D_p$ = Migration distance of particle on picture
$D_r$ = Real migration distance of particle

The result is taken from the average migration distance of 20 particles.

The preferred migration speed of the particles within the gel is preferably achieved by at least one of gel viscosity, gel density and particle density.

The gel preferably has a viscosity of greater than 4000 mPas, preferably greater than 6000 mPas, most preferably more than 10000 mPas. The viscosity was measured with a Brookfield RVT, spindle 27, 2.5rpm at 25°C.

In order to achieve this viscosity the gel preferably contains a thickening agent. A preferred example of a thickening agent is polyacrylic acid.

The gel preferably has a density of more than 1.1 g/cm$^3$, more preferably more than 1.2 g/cm$^3$ and most preferably more than 1.4 g/cm$^3$.

The gel is preferably transparent. Transparent in this context means that particles which are covered by a gel layer of 1cm are still visible under normal daylight conditions.

The gel is preferably substantially water free (having a water content of less than 5%).

Alternatively the gel may have a higher water content with a high ionic strength to prevent the particles from deteriorating in storage. Preferably the water content of the gel is from 5 to 65%, more preferably from 20 to 60% and most preferably from 35 to 50%; the high ionic strength is preferably provided by a salt content which comprises at least 70%, more preferably at least 80% and most preferably at least 90% of the solid content (the non-aqueous component) of the gel.

Preferred examples of salts include phosphates, (such as tripolyphosphates) citrates and sulphates. Most preferably the salts are alkali metal salts, especially sodium and potassium.

The gel composition may comprise a plurality of enzymes. A portion of each enzyme may be disposed within the water-soluble particles.

An advantage of the present invention is that it allows formulation of a detergent gel composition containing two or more antagonistic enzymes. In this context antagonistic implies that one enzyme would upon contact ordinarily cause / be involved in the deterioration of one or more other enzymes present in the detergent gel, possibly together with itself.

This may be achieved by separate encapsulation of one or more of the enzymes within particles in the detergent gel. Namely, (explained with reference to a 2-enzyme containing system) two options are available. In the first option each enzyme may be encapsulated so that whilst the particles are intact interaction of the two enzymes is not possible. In the alternative, only one of the enzymes need be encapsulated to prevent contact.

In the case where a first enzyme is deteriorated by a second it is preferably to contain the first enzyme in the detergent gel and the second enzyme within the particles. In this arrangement the first susceptible enzyme has an opportunity in use after release to carry out its function, before the second enzyme is released from the water-soluble

particle, i.e. before the second enzyme is able to detrimentally affect the first enzyme.

For example starch digesting enzymes such as amylase are usually deteriorated by protein digesting enzymes (proteases) on long-term storage. To address this problem, and using the present invention the amylase may be contained in the gel and the protease within the particles. This concept could of course also be applied in the reverse, wherein the protease is in the gel and amylase is in the particles.

Furthermore due to the nature of the gel an enzyme released prematurely from, for example, a leaking particle is kinetically hindered by the viscous nature of the gel. Thus destructive interaction with its antagonist is at least partially hindered.

The composition preferably comprises an amylase and / or a protease, to aid soil removal. Any of the encapsulation scenarios described in the paragraphs above is contemplated when both enzymes are present.

To further enhance the stability of the encapsulated enzyme a stabilising aid may be present.

Without wishing to be bound by theory it is proposed that the stabilising aid enhances the stability of the enzyme by "blocking" the active site thereof whilst the enzyme is encapsulated in the particle. As soon as the enzyme is dispersed in use (e.g. in a wash liquor) the stabilising aid is most preferably dispersed in the liquor. Thus the active site of the enzyme is free to act.

Preferred examples of stabilising aids include calcium salts, sugars and starches.

The particles are non-soluble in the gel during storage but disintegrate when the gel is exposed to the conditions of a laundry or dishwashing process. A typical dilution of the gel containing such particles in such process is 15-200 g, more preferred 20-

150g most preferred 25-50g of gel in a wash water amount of 4-15L, more preferred 4-8 L.

The particles comprise a water-soluble encapsulating agent. Water-soluble is herein defined when greater than 90% of 1g of such material (in granular form having a particle size from 50-200μm) dissolves after 40 min in a beaker containing 1 L of de-ionised water at 40°C which is stirred with a stirrer revolving at 200 r.p.m.

The encapsulating agent may comprise a coating for the particles. Alternatively the encapsulating agent may comprise a portion of the core of the particle.

In the first case (where the encapsulating agent is a coating) the encapsulating agent may comprise 2-15% by weight, more preferably 2-10% by weight of the particle.

In the second case (where the encapsulating agent comprises a portion of the core of the particle) it is preferred that the encapsulating agent defines a matrix, within which any other components of the particle may be disposed. In this case the encapsulating agent may comprise at least 10% by weight and more preferably at least 20% by weight of the particle.

Most preferably the encapsulating agent comprises a coating.

Preferably the particles comprise a UV absorbing substance. Most preferably the UV absorbing substance is contained in the coating of this particle. A preferred example of a UV absorbing substance is Titanium Dioxide ($TiO_2$)

The encapsulating agent may contain a plasticiser. Preferred plasticisers include polyglycols and non-ionic surfactants.

Preferably the encapsulating agent is a cellulose derivative or a polyvinylalcohol derivative or a combination thereof.

The preferred density of the particles is expressed relative to that of the gel. The gel and the particles have a preferred difference in density no greater than 0.9 $g/cm^3$, more preferably no greater than 0.6 $g/cm^3$ and most preferably no greater than 0.3 $g/cm^3$.

In order to achieve the desired density difference between the gel and the particles, the particles may incorporate a density aid. Preferred examples of density aids include titanium dioxide and calcium salts.

As pure enzymes typically have a dark brown colour, which is usually not appealing to a consumer, a pigment or a dye is generally included in the particles to make them more aesthetically appealing. Preferred examples of pigments include titanium dioxide and calcium salts (both of which provide a white coloration).

As can be seen it has been found that titanium dioxide and / or calcium salts can play a multiple number of roles in the particles (including stabilising agent, density aid and pigment).

The particles have a granule size distribution in which more than 80% of the particles are of the particle size from 50-1000μm, more preferably from 200-800μm and most preferably from 400-700μm.

The particles preferably have a spherical shape. Most preferably the particles are dispersed evenly throughout the gel composition. When being dispersed, it will be appreciated that low shear methods are employed.

The particles may contain other detergent constituents, which are non-aggressive to the enzyme, such as a citrate or a phosphate (e.g. sodium or potassium tripolyphosphate) salt.

Preferably the particles comprise 0.1 to 5.0 weight %, more preferably 0.3 to 3.0 weight % and most preferably 0.5 to 2.0 weight % of the detergent composition.

The detergent composition is intended for use in dishwashing (both manual and automatic, most preferably automatic) and / or laundry applications.

The product is preferably packed in a water-soluble packaging. Such packaging may be produced by thermoforming of a foil and then sealing of the formed and filled container; vertical form-fill-seal processes or injection moulding of compartments and subsequent filling and closing of such compartments.

The invention is now illustrated with reference to the following non-limiting Examples.

**Examples**

Example 1

Protease (Properase supplied by Genecor) and amylase particles were made using a sugar core material mixed with the enzymes to produce prills. The prills were then coated with hydroxypropylmethylcellulose (alternatively polyvinylalcohol or mixtures of the two were used) which contained plasticiser (polyglycol or a nonionioc surfactant) and pigment dye (e.g. $TiO_2$). The resulting particles were comprised of sugar (40-70%), enzyme (2-20%) film forming water-soluble material (2-10%), plasticiser (1-5%), pigment (0-10%) and dye (0-0.2%). Standard prill making and coating technologies provided by e.g. equipment as produced by Glatt were used to make the particles. The particles produced had a particle size wherein 80% of the particles had a diameter in the range 200-600 µm.

A gel was made up having the following composition:

| Component | Wt% |
|---|---|
| Dehardened Water | 37.428 |
| Sulphuric acid (50%) * | 0.105 |
| Dye | 0.0025 |
| Polyacrylic acid (thickener) | 0.800 |
| Sodium citrate | 30.000 |
| Potassium tripolyphosphate | 30.000 |
| Properase particle | 1.080 |
| Amylase particle | 0.340 |
| Perfume | 0.300 |
| | 100.00 |

* Sulphuric acid is added to water to facilitate the dispersion of the thickener. After dispersion of thickener all other ingredients are added.

The resultant detergent composition was stored in sealed glass containers in the dark for twelve weeks at 20°C or 35°C.

Table 1 shows the activity of the enzymes from the particles compared to enzyme activity of an enzyme solution stored under the same conditions.

Table I

| Remaining Enzyme Activity After 12 Weeks | | | |
|---|---|---|---|
| **Protease** | | **Amylase** | |
| Enzyme Particle | | Enzyme Particle | |
| 20°C | 100% | 20°C | 100% |
| 35°C | 89.10% | 35°C | 96.40% |
| | | | |
| Enzyme Solution ** | | Enzyme Solution ** | |
| 20°C | 99.40% | 20°C | 73.40% |
| 35°C | 77.20% | 35°C | 67.50% |

** enzyme solution is available as Purastar ST 15000 L (amylase) and Properase 1600 L (protease) both ex Genencor.

The formulation according to Example 1 shows improved stability of the enzymes when added in the form of particles as described in the specification.

Example 2

The table shows a composition according to the invention.

| Component | Wt% |
|---|---|
| Dehardened water | 61.339 |
| Monopropylene glycol | 1.850 |
| Sulphuric acid (30%) | 0.100 |
| Preservative | 0.100 |
| Polyacrylic acid (thickener) | 1.250 |
| Trisodium citrate | 32.800 |
| Sodium hydroxide | 0.110 |
| Calcium chloride | 0.500 |
| Amylase particles | 0.500 |
| Nonionic surfactant | 0.200 |
| Perfume | 0.050 |
| Protease liquid | 0.650 |
| Amylase liquid | 0.550 |
| Dye | 0.001 |
| | 100.000 |

The formulation according to example 2 shows good stability of the enzyme (amylase) which is contained in the particles.

The detergent composition was stored in sealed glass containers in the dark for twelve weeks at 20°C or 35°C.

Table II shows the activity of the enzyme from the particles compared to enzyme activity of an enzyme solution stored under the same conditions.

Table II

| Remaining Enzyme Activity After 12 Weeks | |
|---|---|
| Amylase | |
| Enzyme Particles | |
| 20°C 100% | |
| 35°C 95% | |
| Enzyme Solution ** | |
| 20°C 73.40% | |
| 35°C 67.50% | |

** enzyme solution available as Purastar ST 15000L (amylase)

## CLAIMS

1. A detergent composition comprising an enzyme which is at least partially encapsulated within water-soluble particles in a gel, the particles comprising a water-soluble encapsulating agent, wherein the particles have a migration speed in the gel of less than one centimetre per month.

2. A composition according to claim one, wherein the migration speed of the particles is less than 0.7 cm per month.

3. A composition according to claim 1 or 2, wherein the migration speed of the particles is less than 0.4 cm per month.

4. A composition according to claim 1, 2 or 3, wherein the composition has a viscosity greater than 4,000 mPas, more preferably greater than 6,000 mPas and most preferably greater than 10,000 mPas.

5. A composition according to any one of claims 1 to 4, wherein the gel contains a thickening agent.

6. A composition according to claim 5, wherein the thickening agent is polyacrylic acid.

7. A composition according to any one of claims 1 to 6, wherein the composition has a density of greater than 1.1 $g/cm^3$, more preferably greater than 1.2 $g / cm^3$ and most preferably greater than 1.4 $g / cm^3$.

8. A composition according to any one of claims 1 to 7, wherein the gel is substantially water free having a water content of less than 5%.

9. A composition according to any one of claims 1 to 7, wherein the gel has a water content of from 5 to 65%, more preferably from 20 to 60% and most preferably from 35 to 50%.

10. A composition according to claim 9, wherein the composition has a salt content of at least 70%, more preferably at least 80% and more preferably at least 90%.

11. A composition according to claim 10, wherein the salt is a phosphate, citrate or sulphate salt.

12. A composition according to any one of claims 1 to 11, wherein the composition comprises a plurality of enzymes.

13. A composition according to any one of claims 1 to 12, wherein the particles contain a stabilising aid for the enzyme.

14. A composition in accordance with any one of claims 1 to 13, wherein the encapsulating agent comprises a coating for the particles.

15. A composition in accordance with claim 14, wherein the encapsulating agent comprises 2 to 15% by weight of the particle, more preferably 2 to 10% by weight of the particle.

16. A composition in accordance with any one of claims 1 to 13, wherein the encapsulating agent comprises a portion of the core of the particle.

17. A composition in accordance with claim 16, wherein the encapsulating agent comprises at least 10% by weight and more preferably at least 20% by weight of the particles.

18. A composition in accordance with any one of claims 1 to 17, wherein the particles comprise a UV absorbing substance.

19. A composition in accordance with claim 18, wherein the UV absorbing substance is titanium dioxide.

20. A composition in accordance with any one of claims 1 to 19, wherein the encapsulating agent contains a plasticiser.

21. A composition in accordance with claim 20, wherein the plasticiser is a polyglycol or a nonionic surfactant.

22. A composition in accordance with claim 20 or 21, wherein the encapsulating agent is a cellulose derivative, a polyvinyl alcohol or an admixture thereof.

23. A composition in accordance with any one of claims 1 to 22, wherein the gel and the particles have a difference in density of no greater than 0.9 g / $cm^3$, more preferably no greater than 0.6 g / $cm^3$ and most preferably no greater than 0.3 g / $cm^3$.

24. A composition in accordance with claim 23, wherein the particles contain a density aid.

25. A composition in accordance with any one of claims 1 to 24, wherein the particles contain a dye / a pigment.

26. A composition in accordance with any one of claims 1 to 25, wherein more than 80% of the particles have a particle size from 50 to 1,000 micrometres, more preferably from 200 to 800 micrometres and most preferably from 400 to 700 micrometres.

27. A composition in accordance with any one of claims 1 to 26 for use in dishwashing and / or laundry applications.

28. A composition in accordance with any one of claims 1 to 27, disposed in a polyvinylalcohol packaging.

29. A method of dishwashing and / or laundry comprising the use of a detergent composition in accordance with any one of claims 1 to 26.

## Abstract

The present invention comprises a detergent composition comprising an enzyme. The enzyme is at least partially encapsulated within water-soluble particles in a gel. The particles comprise a water-soluble encapsulating agent. The particles have a migration speed in the gel of less than one centimetre per month.